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Preparation and Characterization of Nanostructured Manganese-Doped Zinc Oxide Thin Films



Abstract: - By spray pyrolysis (SP) process, thin films of ZnO and (ZnO:Mn) with 1% and 3% concentrations were created at a temperature of 400 °C. According to XRD investigation, ZnO films are polycrystalline and have a cubic structure with a distinct peak in one direction (101). The grain size increases as manganese content rise, from 12.66 nm to 14.66 nm. While the strain (ϵ) for ZnO reduced after manganese doping, it decreased from 27.36 to 23.63. Surface topography and nanostructure study reveal that as the manganese (Mn) content of ZnO films increased, cluster grain size, average roughness, and root mean square roughness (Rrms) all significantly reduced. The average transmittance was >70% in the visible area for Undoped ZnO and 1, 3% Manganese doping optical transmittance demonstrates exceptional optical transparency. When doping levels are increased by 1% or 3%, the absorption coefficient rises. The optical band gap was decreased from (3.32 to 3.21) eV. Results illustrate that the films' refractive index and extinction coefficient decreases with increasing Tin Doped.

Keywords: ZnO, Mn, XRD, topography, transmittance, absorption coefficient, band gap.

I. INTRODUCTION

Zinc oxide has many applications due to its excellent optical, photoelectric, and piezoelectric properties. Thus, this approach is ideal for realizing optical devices, including LED, and solar cells. In the applications above, zinc oxide (ZnO) with a hexagonal wurtzite structure has recently shown potential as an ITO alternative [1-4]. It is particularly interested in wide bandgap semiconductors due to the growing commercial need for short-wavelength LED [5-8]. ZnO-nanostructured films are a great option because of their wide bandgap (3.37 eV) [8, 9]. ZnO is grown using a variety of techniques, such as CVD [10, 11], sol-gel [12-14], magnetron sputtering [15, 16], laser molecular beam epitaxy [17] and as spray pyrolysis (SP) [18-24]. Spray pyrolysis is one of these techniques suitable for broad-area thin film production and simple doping. We report the experimental findings of physical attributes discovered using various characterization methods in this paper. The thin films are grown via SP technique. The temperature and deposition time are crucial because this is a chemical procedure. The Manganese

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Doping significantly impacts the deposited Undoped ZnO thin films. Manganese doping has been used to develop several thin films while maintaining stable substrate temperatures

II. EXPERIMENTAL

The current study used CSP to manufacture Mn-doped ZnO film on a glass slide substrate. Thin films of ZnO were produced using this technique. 0.1 M of ZnCl₂ dissolved in the 1:1 mixture of deionized water and ethanol was used to manufacture ZnO thin films. The doping substance was manganese trichloride (MnCl₃), which was dissolved in deionized water and then had a few drops of hydrochloric acid added to it so the solution would be transparent. The following are the requirements for preparation: Base temperature was 400 °C, there was a distance of 28 cm between spout and the substrate, and the spraying period was 8 S , but it was prolonged by 65 seconds to prevent cooling, the spray rate was 5 ml/min. and N₂ was employed as the transporter gas. The thickness was determined using the gravimetric method of 340 ± 20 nm. ZnO thin film generated was established by XRD, and AFM was used to evaluate the films' structure and morphology. Transmittance is estimated using a UV-Vis NIR.

III. RESULTS AND DISCUSSIONS

Figure (1) displays the XRD patterns of ZnO thin film created using a straightforward chemical method before annealing. This figure has several peaks at angles of 31.73°, 36.24°, 47.42°, and 62.65° that are attributed to the (100), (101), (102), and (103) planes, respectively. These peaks could be fitted by ICDD card number (36.1451) [25, 26], showing the presence of polycrystallinity and predominant peak in (101) plane [27, 28].

The grain size (*D*) was calculated using Scherrer's Eq. (1) [29-31]:

$$D = \frac{0.9\lambda}{\beta \cos\theta} \quad (1)$$

Where λ is the X-rays' wavelength, β and θ are FWHM and the Bragg angle of (101) peak, respectively. Table 1 displays the outcomes that were attained. The findings demonstrate that *D* rises from 12.66 to 14.66 nm when the ZnO content is increased to ZnO: 3% Mn. As a result, the manganese content is crucial in adjusting the crystal sizes of the material [32, 33].

Evaluation is also done on other structural metrics, including dislocation density (δ) from Eq. 1[34-36].

$$\delta = \frac{1}{D^2} \quad (2)$$

The calculation of the strain (ϵ) is calculated via Eq. 2 [37-39]:

$$\epsilon = \frac{\beta \cos\theta}{4} \quad (3)$$

Table 1 displays the data, and it is clear that the strain declines as the manganese content rises. The regular atom arrangements in the crystal lattice are responsible for improving crystalline quality [15]. We can infer from these findings that the manganese concentration fundamentally impacts crystallite size and decreases from 12.66 to 14.66 [40,41]. Figure (2), shows the structural parameters S_{para} as functions of Manganese concentration.

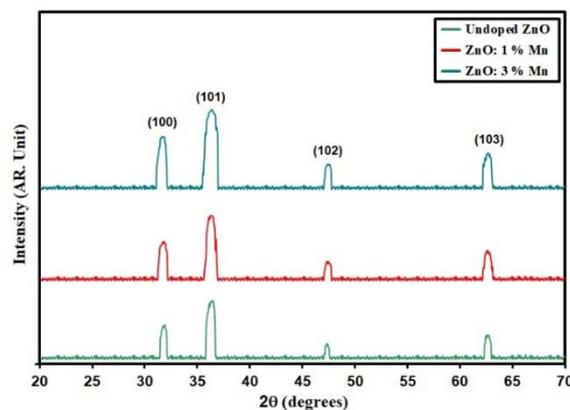


Fig.1. XRD patterns.

TABLE 1. D , E_g and S_{para} of prepared films.

Samples	2θ ($^\circ$)	(hkl) Plane	β ($^\circ$)	E_g (eV)	D (nm)	$(\delta) (\times 10^{14})$ (lines/m 2)	(ϵ) ($\times 10^{-4}$)
Undoped ZnO	36.24	101	0.66	3.32	12.66	62.33	27.36
ZnO: 1% Mn	36.21	101	0.62	3.26	13.48	54.97	25.7
ZnO: 3% Mn	36.17	101	0.57	3.21	14.66	46.47	23.63

The three-dimensional surface morphology is depicted in Fig. 3. Table 2 displays the produced films' average particle size P_{av} , root mean square roughness (R_{rms}), and average roughness (R_a). ZnO, ZnO:1% Mn, and ZnO:3% Mn nanoparticles were found to have average particle sizes in the range of (92.6), (85.7), and (47.8) nm, respectively. When doping was reduced, the R_{rms} value is dropped to 1.99 nm [42, 43]. R_a values are shown in Figs. 3(a3), (b3), and (c3), respectively. The AFM parameter P_{AFM} values are shown in Table (2).

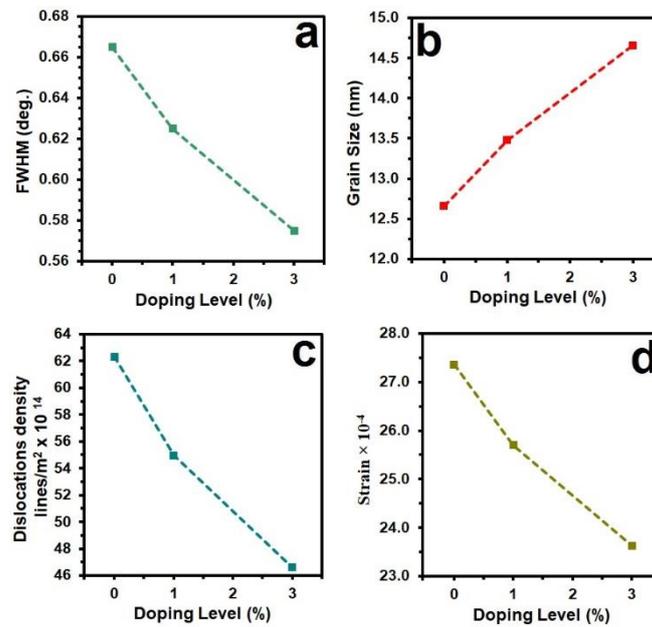


Fig.2. S_{para} of the grown films.

TABLE 2. P_{AFM} of the intended films.

Samples	P_{av} nm	R_a (nm)	R_{rms} (nm)
Undoped ZnO	92.6	8.31	10.81
ZnO: 1% Mn	85.7	6.27	6.15
ZnO: 3% Mn	47.8	3.96	5.23

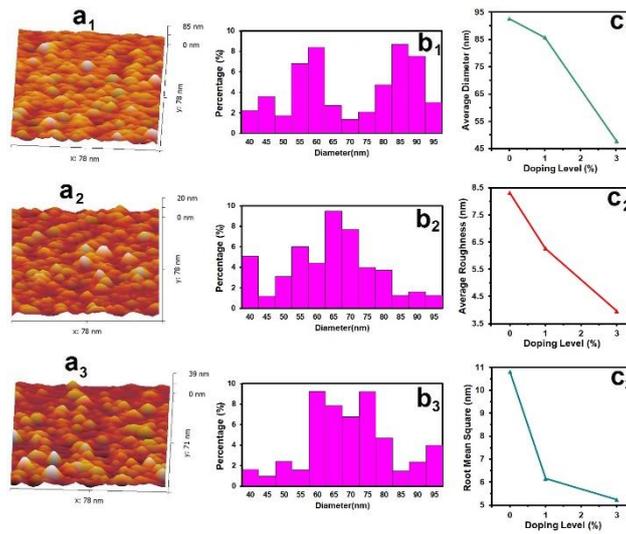


Fig. 3. AFM information's.

Fig. (4) offers the transmittance (T) spectra of ZnO film and doped films. The transmittance of ZnO film decreases by manganese content [44, 45].

The absorption coefficient (α) is determined [46-48]:

$$\alpha = \frac{2.303A}{t} \quad (4)$$

Where t is the film thickness, Fig. 5 depicts α increases with ncrease of manganese doping [49, 50].

The energy gap E_g is plotted following Tauc relation [51-53]:

$$(\alpha h\nu) = A(h\nu - E_g)^{\frac{1}{2}} \quad (5)$$

Where A is the constant, ν is the incoming radiation's frequency, and h is Planck's constant; Figure 4 shows the energy gap values were 3.32 and 3.21 eV, respectively. This graph demonstrates how the energy gap value of manganese-doped ZnO film is decreasing. These ideals align well with those expressed by other employees [54, 55].

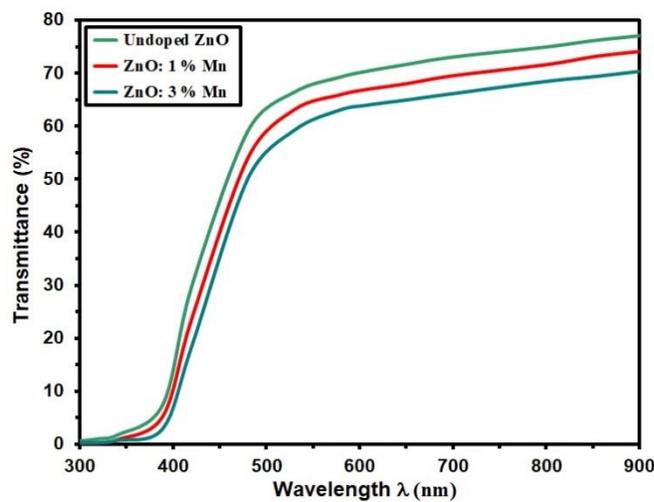


Fig. 4 T versus wavelength of the grown films.

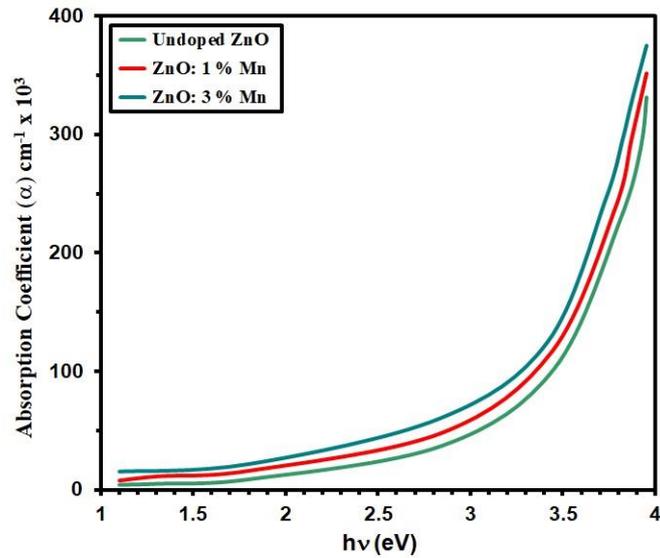


Fig. 5 α versus $h\nu$ for deposited films.

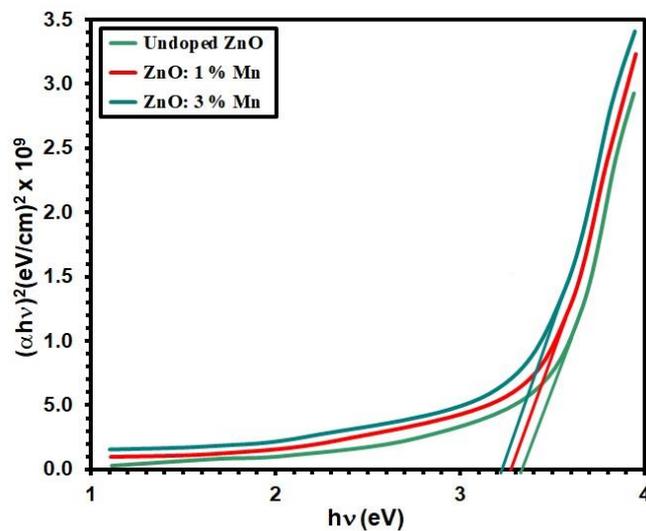


Fig. 6 E_g of the grown films.

The extinction coefficient (k) is measured by Eq. 6 [56-58]:

$$k = \frac{\alpha\lambda}{4\pi} \quad (7)$$

The variation of k is seen in Fig. 7. After magnesium doping, there is a modest drop in k , which is related to the wavelength of polarized light. k immediately recaptures its characteristic of absorption [59,60]. The refractive index n is obtained by Eq. (7) [61-63]:

$$n = \left(\frac{1+R}{1-R} \right) + \sqrt{\frac{4R}{(1-R)^2} - k^2} \quad (8)$$

Where R is the reflectance, Fig. 8. displays n spectr against λ . There is a slight decrease in refractive index via Mn content [64, 65].

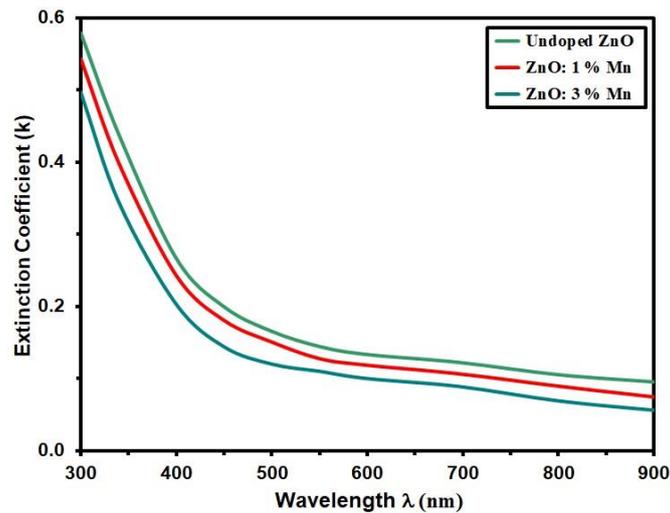


Fig. 7 k of the grown films.

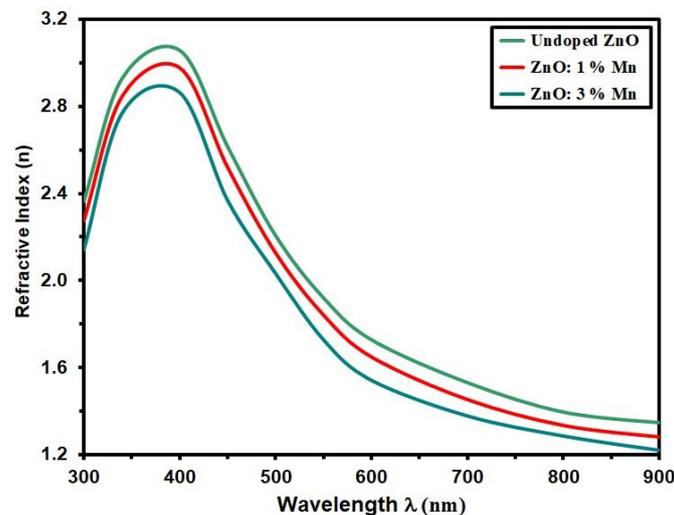


Fig. 8 n for grown films.

IV. CONCLUSION

We looked at spray pyrolysis-deposited thin films of zinc oxide. According to XRD findings, annealed ZnO films have a preferred orientation of 101 degrees. For prepared samples, microstructural parameters have been computed. Particle size increases as the manganese content does, from 12.66 nm to 14.66 nm. Undoped ZnO, ZnO:1% Mn, and ZnO:3% Mn nanoparticles were found to have grains between 92.6, 85.7, and 47.8 nm in size, respectively. The transmittance in the visible range is greater than 78 %. The absorption coefficient decreased by increase of Mn content and the bandgap values decreased from 3.32 to 3.21 eV with increasing of Manganese doped zinc oxide. Extinction coefficient and refractive index are lowered with manganese concentration.

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